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SPRAY COMBUSTION MODELING

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1 Introduction

Concern over the future availability of high quality liquid fuels for use in furnaces and boilers prompted the U. S. Department of Energy (DOE) to consider alternate fuels as replacements for the high grade liquid fuels used in the 1970's and 1980's. Alternate fuels were defined to be combinations of a large percentage of viscous, low volatility fuels resulting from the low end of distillation mixed with a small percentage of relatively low viscosity, high volatility fuels yielded by the high end of distillation. The addition of high volatility fuels was meant to promote desirable characteristics to a fuel that would otherwise be difficult to atomize and burn and whose combustion would yield a high amount of pollutants. Several questions thus needed to be answered before alternate fuels became commercially viable. These questions were related to fuel atomization, evaporation, ignition, combustion and pollutant formation

The Jet Propulsion Laboratory (JPL) has been funded by DOE for several years since the 1980's to investigate evaporation, ignition, combustion and aspects of pollutant control. The initial part of this investigation has been performed under the Energy Conservation and Utilization Technologies (ECUT) program and focussed on the prediction of evaporation of drops in sprays including the crucial aspect of drop interaction, on the characterization of cenospheres (carbonaceous, porous particles necessarily resulting from the combustion of alternate fuels), and on the combustion of these cenospheres as a way to extract additional energy out of the fuel while diminishing the mass of pollutants. The results from these studies have been summarized in previous reports. These results have shown, in agreement with previously unexplained experimental observations, that the parameters controlling the evaporation of

drops in the dense regime, where the drops are so closely spaced that they influence each other, are qualitatively different from those in the dilute regime where the drops act as if they were isolated. Moreover, it also has been shown that there are more control parameters in the dense regime than in the dilute regime. Based upon this result, it has been suggested that spray control should be planned in the dense regime, close to the atomizer. This suggestion is in agreement with well-known engineering practice.

After a hiatus of two years, JPL has been asked by the Advanced Industrial Concepts Division and further by the Industrial Energy Efficiency Division to conclude this study by investigating the ignition and combustion aspects of alternate fuels. This Final Report describes briefly the most salient findings of this study. The details of the study can be found in the three appendices of this report which are papers that have been submitted for publication in technical journals and/or presentation at technical meetings.

It should be noted that the present study pertaining to the behavior of alternate fuels, which are multicomponent fuels, is pertinent not only to the particular study undertaken here, but has a more general range of applicability. This is because practically all liquid fuels that are industrially burned are multicomponent fuels. Thus, the results found under this investigation are qualitatively applicable to the aviation and automotive industries.

2 Ignition of a binary-fuel cluster of drops

2.1 General discussion

Similar to the previous study on evaporation, a cluster of drops has been chosen as a representative element of a spray, and a binary fuel (a fuel composed of two pure compounds) has been chosen to represent an alternate fuel. The first choice has been motivated by experimental observations showing that drops cluster both in combusting and non-combusting sprays; in combusting sprays flames have been seen to enclose groups of drops thereby showing the important effect of drop interaction. Without this interaction, flames will surround each individual drop as predicted by the classical isolated-drop theory. The second choice has been dictated by industrial considerations: industrially used liquid fuels may be mixtures of tens of pure compounds making it cumbersome to account for all of them in calculations which may be performed on a workstation rather than on a supercomputer. Thus, a binary fuel is considered to be composed of a large percentage of a viscous, low volatility fuel—the solvent—and a low percentage of a low viscosity, high volatility fuel—the solute. It is envisaged that in an industrial setting each of the two components will have the average (by initial composition mass fractions) characteristics of respectively the low end distillation compounds and the high end distillation compounds. One limiting aspect for making this partition into two components is the fact that the concept of

volatility is not thermodynamically well defined; it is an engineering term related to the evaporation of a liquid from a flat surface so that in fact it is related to both the latent heat of evaporation and to the saturation vapor pressure versus the temperature. However, when two fuels are compared (such as n-decane and n-hexane, for example), one might have a larger latent heat (n-hexane) which means that it is more difficult to evaporate, while it also has a higher saturation pressure which means that it is easier to evaporate. The question is: which one is more volatile?

This question was answered by the results of the ignition study which is presented in Appendix 1 and whose highlights are also given in Appendix 3. Other important questions related to the influence of the solvent and solute identity, to the initial mass fraction of solute in the fuel, to the effect of the initial surrounding gas temperature, to the effect of the initial slip velocity between drops and gas, to the effect of the initial cluster size and to the effect of the chemical kinetics of the fuel were answered as well.

The study has been performed under the assumptions that the solute has a volatility much higher than the solvent and that the kinetics of the two components are independent. The first assumption is justified by the choice of the compounds whose properties are averaged to yield the solvent and the solute as conceptualized above. The second assumption is justified by the lack of chemical kinetic data on all but the simplest three hydrocarbons; it is recognized that the present results might change when future such data is incorporated into the model.

2.2 Highlights of the results

Calculations have been performed by varying the air/fuel mass ratio for different values of the main variables. Small values of the air/fuel mass ratio correspond to very dense clusters of drops, whereas large values of the air/fuel mass ratio correspond to very dilute clusters of drops in which there is no drop interaction.

Baseline results show that in a typical near-field-atomizer gas temperature, ignition is strongly controlled by the solvent in the very dense cluster regime, and strongly controlled by the solute in the very dilute regime. In the intermediary dense and dilute regimes, control by either one of the components is very weak and might depend among other variables upon the detailed kinetic interaction that is presently unknown. Thus, the isolated drop results are recovered in the very dilute regime; however, it is found that it is qualitatively a different process which controls ignition in a real spray. The practical conclusion is that at typical near-field-atomizer gas temperatures it is useless to use more expensive fuels having larger amounts of high grade compounds for the purpose of enhancing ignition because it is only the very dilute clusters of drops which will benefit from this treatment. These clusters of drops appear only at relatively large distances from the atomizer and thus do not participate in the ignition process.

As the surrounding gas temperature is increased, it is easier to gain control of

ignition over a larger range of air/fuel mass ratios. This is achieved by using the solute in the very dilute and increasingly in the dilute regime, and by using the solvent in the very dense and increasingly in the dense regime. Eventually, a temperature is reached at which ignition control of the entire range of air/fuel mass ratios, and thus of the entire spray, can be achieved.

Studies of the effect of solvent identity revealed that the concept of volatility is associated more with the latent heat of evaporation in the very dense cluster regime (because of the competition among drops for available heat), and it is associated more with the saturation pressure in the very dilute cluster regime (because heating of the drop is no longer an issue and it is the surrounding vapor pressure which control evaporation).

It has also been shown that variations of the initial slip velocity between drops and gas affect results only in the very dilute regime because of the short relaxation time of the slip velocity in the very dense regime. In contrast, changes in the size of the cluster affect the ignition timing only in the very dense regime which is limited by the amount of heat available to each drop to increase its temperature so that it may evaporate. A smaller cluster size corresponds to a larger cluster-surface area per cluster volume, and thus heat transfer to the cluster is enhanced.

In the very dilute cluster regime and for very low values of the initial solute mass fraction, ignition is found to occur around groups of drops inside the cluster. For all other conditions, ignition occurred around the entire cluster in agreement with experimental observations of burning sprays.

All of the above results were found under the assumption that the chemical kinetic parameters of solvent and solute were identical. Studies of the effect of the chemical kinetics show that it has an overwhelming effect on the ignition results. Solvent-controlled ignition can be lost when the solvent activation energy is slightly increased. Similarly, solute-controlled ignition can be lost when the solute activation energy is slightly increased. Since chemical kinetic parameters for hydrocarbons more complicated than propane are unknown, definitive conclusions must await precise chemical kinetic information.

3 Cluster combustion of binary-fuel drops

3.1 General discussion

The study of cluster combustion was motivated by the experimental evidence of flames surrounding clusters of drops. JPL performed under DOE-ECUT sponsorship a study of single-component (a single pure fuel) cluster combustion. This previous model documented in a technical publication has been now extended to account for the combustion of two components. One conclusion of the previous study is that cluster flames exist only in a restricted range of air/fuel mass ratios. This is because if this ratio is very small, the cluster might be so dense that the drops extract too

much heat from the gas during evaporation, before heat transfer processes from the cluster surroundings may replenish it, and thus the temperature becomes too low to initiate ignition. For these clusters, evaporation proceeds without ignition until the drops disappear. Ignition might occur later in the gas phase, but this situation is outside the scope of this study. When the air/fuel mass ratio is very large at ignition, the gas mixture inside the cluster is fuel-lean and internal cluster combustion (which necessarily occurs before the flame migrates outside the cluster) depletes all gaseous fuel inside the cluster. With no gaseous fuel left to escape the cluster, the external cluster flame cannot become established. These two situations represent the lower and upper limits for the existence of cluster flames. Thus, cluster flames exist for clusters which are not too dense and for which the gaseous mixture is fuel-rich at ignition.

The combustion model describes a quasi-steady flame following ignition of either solvent or solute. The evaporation and ignition model are those described in the previous section. As mentioned above, a flash flame sweeps the interior of the cluster before the flame migrates outside the cluster. During flash-flame combustion, oxygen is apportioned between solvent and solute in proportion to their average mass fraction at ignition. It is the igniting component which determines the flame position outside the cluster through the solution of the conservation equations.

Results from this study are presented in detail in Appendices 2 and 3. What is presented below are highlights of the findings.

3.2 Highlights of the results

An extensive study has been performed by varying solvent and solute identities, the initial drop velocity, the initial mass fraction of the solute and the activation energy.

The results show that stronger flames that are established further from the cluster surface occur for smaller initial air/fuel mass ratios (corresponding to denser clusters) and larger initial drop velocities. In this case, it is only a small amount of fuel that is burned before drop disappearance, with the remaining fuel to be burnt at the rate prevailing in the surrounding gas. Weak flames are established extremely close to the cluster surface; they occur mainly for large initial air/fuel mass ratios and small initial drop velocities. These flames behave asymptotically (as the air/fuel mass ratio reaches the upper limit for which cluster flames exist) like classical quasi-steady diffusion flames where the fuel emitted by the cluster is almost entirely burnt in the flame. For intermediary values of the initial drop velocity, the classical behavior of the diffusion flame is never reached, indicating the importance of convective effects.

For diffusion-dominated combustion, the fraction of fuel burnt is an increasing function of the initial air/fuel mass ratio because ignition occurs earlier during the drop lifetime. As convective effects become important, the flame is relatively stronger in the small air/fuel mass ratio regime than in the purely diffusion regime and as a result it burns a larger fraction of fuel. Thus, for intermediary convective combustion,

the fuel mass fraction burned is a nonmonotonic convex function of the initial air/fuel mass ratio. When convection dominates, the flame is considerably stronger for small initial air/fuel mass ratios and accordingly an increasing fuel fraction is burned. The total fraction of fuel burned (flash flame and external combustion) is an increasing function of the initial air/fuel mass ratio since the later ignition for small values of this ratio also corresponds to situations where there is less oxygen inside the cluster and thus less fuel may be consumed by the flash flame.

Changing the ignition kinetics translates and enlarges or shrinks the collective flame regime as a function of the initial air/fuel mass ratio, but does not change qualitatively the results.

4 Summary and conclusions

This study has focussed on some critical aspects of liquid fuel evaporation, ignition and combustion: the effect of drop interactions coupled to the effect of real-fuel composition. Results show that near the atomizer, where drop interactions are important, there are more drivers than in the regions farther from the atomizer, and that these drivers are qualitatively different from those found in regions far from the atomizer. The conclusion is that spray control and optimization should be planned near the atomizer, and that it is imperative to utilize knowledge derived from interactive-drop calculations in order to insure that the chosen driver is indeed effective in this regime.

A APPENDIX 1

Paper submitted for publication and now in the revision process

B APPENDIX 2

Paper to be presented at the conference of the Institute of Liquid Atomization and
Spray Systems, 5/21-24, 1995

C APPENDIX 3

Paper to be presented at the 2nd International Symposium on Liquid Rocket
Propulsion, 6/19-21, 1995

CAN ONE CONTROL IGNITION OF A BINARY-FUEL SPRAY?

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ABSTRACT

Evaporation and ignition of a binary-fuel cluster of drops is described by models under the assumptions that the volatile compound has infinite volatility with respect to the solvent and that the chemistries of the two compounds are independent. A Damkohler number criterion developed for use in sprays is utilized to determine the ignition time. Another criterion is used to determine the ignition location which can be either around individual drops, or around groups of drops inside the cluster, or around the entire cluster.

Results show that except for very dilute situations where the initial liquid mass fraction of the volatile is very small, ignition always occurs around the entire cluster. Otherwise, ignition occurs around groups of drops inside the cluster but never around individual drops even though the ratio of the distance between the centers of two adjacent drops by the drop diameter is greater than thirty five.

Parametric studies performed by varying the air/fuel mass ratio for a variety of parametric combinations show that : (1) At typical gas temperatures for combustion devices, the ignition of very dense clusters of drops is strongly-controlled by solvent ignition, whereas that of very dilute clusters of drops is strongly-controlled by ignition of the volatile. In the dense and dilute regimes neither the solvent nor the volatile strongly control ignition. These conclusions are independent of the amount of volatile initially present in the liquid. (2) The concept of volatile is more strongly associated with the latent heat of evaporation in the very dense regime, and more strongly associated with the saturation pressure curve in the very dilute regime. (3) By increasing the ambient gas temperature one gradually gains control of ignition in the dense and dilute regimes through the solvent and volatile respectively. (4) The initial slip velocity between phases affects ignition only in the very dilute regime. (5) Changes in the cluster size affect only the ignition time in the very dense regime. (6) The previous conclusions are valid under the assumption of identical kinetics for the two compounds. When different kinetics is considered, it turns out that kinetic effects overwhelmingly dominate ignition.

INTRODUCTION

The majority of practical fuels used either in power plants or for propulsion are multicomponent fuels. This means that they are composed of several chemical species, and that, in principle, the interaction among the various characteristics of the pure chemical components determines the characteristics of the fuel in terms of evaporation, ignition and combustion.

Since many of these multicomponent fuels contain tens and sometimes more than a hundred pure components, it is found convenient to categorize these components according to a criterion which is thermodynamically not well defined, but which is engineeringly useful; this is the concept of volatility. Volatility is, a priori, thermodynamically related both to the latent heat of evaporation and to the saturation vapor pressure curve versus the temperature. Using the concept of volatility, the multitude of chemical components is partitioned into two components: the solute and the volatile. The solute contains the heavier, more viscous components which evaporate slowly. The volatile contains the lighter, less viscous components which evaporate rapidly. When these criteria are used to define solute and volatile respectively, it turns out that the concept of volatility is more strongly related to the saturation vapor pressure curve versus the temperature in the very dilute region of the spray, and more strongly related to the latent heat of evaporation in the very dense region of the spray as will be shown in this paper.

Single droplets of multicomponent fuels have been studied extensively in the past [1-7]. It was found that isolated drop evaporation in a convective flow is dominated by liquid mass diffusion [1-5] due to the creation of Hill vortices inside the drops through the intermediary of the shear layer which forms at the drop surface as a result of the slip velocity between phases. The internal convection thus established enhances liquid mass diffusion, which is otherwise a

very slow process, to the point that liquid mass diffusion of the volatile becomes a controlling process during evaporation. Studies of isolated drop ignition [6,7] understandably identified the volatile as the component initiating ignition, and it was found that the addition of a small amount of volatile enhances ignition considerably [7]. Mawid and Aggarwal [7] acknowledge that their study is only valid in the dilute limit; an indication of this restriction is the fact that they find ignition to be dominated by individual drop ignition rather than by group - drop ignition. In contrast, experimental observations of burning sprays [8-14] show multiple flames, each flame surrounding a group of drops. Individual drop ignition with flames quickly merging and surrounding groups of drops is still possible, however it has not been experimentally observed.

A more recent study of clusters of drops [15] portraying the groups of drops identified experimentally both in burning [8-12] and non burning sprays [11-14] has revealed a different physical picture than that emerging from the isolated drop studies. The results showed that there are two processes which may influence evaporation of the volatile. The first process is liquid mass diffusion already identified by the isolated drop studies. The second process is surface layer stripping which is the removal of the volatile at the rate of evaporation of the solute.

For dense clusters of drops the slip velocity between drops and gas relaxes very fast [15,16] because the cluster exposes a large area to the flow (there are many drops per unit area). Thus, there is no time to establish a shear layer at the drop surface, and as a consequence internal drop circulation through the Hill vortex does not occur. The result is that liquid mass diffusion remains a slow process with a characteristic time larger than the drop lifetime, and thus cannot influence evaporation of the volatile. The mass fraction of the volatile within the drop stays frozen in time, and the volatile leaves the surface at the same rate as the solute; this is surface

layer stripping. Experiments performed in low pressures so as to suppress buoyancy [5] have shown precisely this type of behavior: the mass fraction of the volatile stays frozen in time and the volatile leaves the surface at the rate of evaporation of the solute.

For dilute clusters of drops it was found [15] that the slip velocity between phases relaxes in a time comparable to the lifetime of the drop, and thus both liquid mass diffusion and surface layer stripping are important. The parameter characterizing the relative importance of these two phenomena, Be , is the ratio of the mass regression rate to a characteristic volatile diffusion rate [15]. If $Be \ll 1$, then diffusion into the drop boundary layer determines the rate of species transfer from the liquid core to the drop surface and liquid mass diffusion is important. If $Be \gg 1$, then surface layer stripping is important. Plots of Be versus the residual drop radius show that for initially dense clusters of drops $Be \gg 1$ during the drop lifetime (after adjustment from the initial condition). In contrast, for initially dilute sprays $Be = [O(1) - O(10)]$ during the drop lifetime (after adjustment from the initial condition).

On the basis of this behavior it has been possible to classify clusters of drops according to the value of the nondimensional radius of the sphere of influence around each drop. For monodisperse clusters, the nondimensional radius of the sphere of influence is defined as the ratio of the drop diameter by the distance between the centers of two adjacent drops. When this ratio has a value smaller than 10, the cluster is very dense. When this ratio has a value between 10 and 15, the cluster is dense. Dilute clusters are those for which the value of the radius of the sphere of influence is larger than 15 but smaller than 30. Very dilute clusters are those for which the nondimensional radius of the sphere of influence is larger than 30. The drops in very dilute clusters behave as if they were isolated from each other.

The present study addresses ignition of clusters of binary - fuel drops composed of a solute and a volatile as discussed above. The model for drop evaporation is that of Harstad and Bellan [15] and it is briefly recalled in the next section. The ignition model is described in the next section as well. Results are obtained for a variety of fuel combinations and an extensive parametric study is performed. It is shown that when the kinetic parameters are identical for the two components it is only in the very dilute regime that ignition is governed by the evaporation of the volatile; this is a situation which does not have practical interest since spray ignition is desirable well before the spray becomes dilute. It is also shown that, for the same kinetic parameters, in the very dense regime ignition is controlled by the solute. Interestingly, for identical kinetic parameters and at a typical ambient gas temperature in combustion devices, ignition cannot be controlled in the dense or dilute regimes because neither solvent or volatile are strongly dominating ignition. Departures from these conclusions are shown to occur when some parameters are varied.

PHYSICAL CONFIGURATION AND MODEL

1. The evaporation model.

The physical configuration studied here is exactly the same as in Harstad and Bellan [15], but whereas in that investigation it is only evaporation that was studied, here ignition may occur as well. As will be discussed below, ignition of a multicomponent fuel is an intricate chemical problem which has not yet received the interest that it deserves because of the lack of precise chemical kinetic information regarding even single-component fuels.

Following the configuration described in Ref. 15, in the present study, a cluster composed of binary-fuel, monodisperse drops moves in an ambient gas with a velocity u_d which is measured with respect to a coordinate system fixed with respect to the ambient. The cluster is assumed to be spatially homogeneous in thermodynamic quantities on a length scale of the order of many drop radii. Initially the gas inside the cluster is quiescent, and so is the gas in the ambient. However, once the drops start moving, the gas inside the cluster acquires a velocity, u_g . Thus, the axial slip velocity between drops and gas inside the cluster is $u_{d,g} = u_d - u_g$.

Since the temperature of the ambient is much larger than the initial drop temperature, the drops heat up, evaporate and might ignite. The pressure inside the cluster is atmospheric and constant, so that the change in the gas temperature inside the cluster implies a change in the volume of the cluster. The boundary of the cluster is by definition the envelope of the drops which moves with a velocity dR/dt with respect to the cluster center. Thus, the drops have not only an axial velocity with respect to the gas, but also a radial velocity. The motion of the drops is assumed to be self-similar, so that drops occupy positions $r = \xi R$, where R is the cluster radius and ξ is fixed for a given drop; $0 < \xi < 1$. With this definition, the relative radial

motion for a drop interior to the cluster is ξu_{ge} , where u_{ge} is the gas radial motion at the cluster edge. The relative radial motion of a drop at the cluster edge is $u_{re} = dR/dt - u_{ge}$. The dynamics of the drops has been described in detail elsewhere [17-19] and thus will not be repeated here. Of special interest is the fact that the drag coefficient depends upon the drop number density, the blowing of mass from the drops as they evaporate (which depends itself upon the drop number density), and the slip velocity inside the cluster.

The binary fuel is assumed to be composed of a solute and a volatile whose volatility is much larger than that of the solute; practically, the volatility of the solute is infinite with respect to that of the volatile [15]. Each drop is composed of a liquid core whose motion is described by the Hill vortex solution, and a thin boundary layer at the drop surface. The evaporation model is described in detail in Ref. 15. The highlight of the model is the definition of a nondimensional quantity, Be , which represents the ratio of the drop mass regression rate to a characteristic volatile diffusion rate. As discussed above, when $Be \ll 1$, diffusion into the drop boundary layer governs the rate of species transfer from the liquid core to the drop surface and evaporation from the surface occurs at a rate defined by the Langmuir - Knudsen evaporation law. In fact it is the slower of these two rates which governs evaporation since the two processes are sequential. When $Be \gg 1$, the transfer of volatile from the liquid core to the gas phase is governed by surface layer stripping, that is by the regression rate of the drop [15]. Because $Be \gg 1$ for drops in dense clusters, whereas $Be = [O(1) - O(10)]$ for drops in dilute clusters [15], a qualitatively different process is found to control evaporation of the volatile in these two regimes.

2. The ignition model.

Ignition is modeled under the assumption that the chemistry of the solute and that of the volatile are independent. This assumption is not realistic for hydrocarbons which decompose into simpler compounds who ultimately interact chemically. However, given the lack of knowledge of the detailed chemistry for hydrocarbons more complicated than propane, any other assumption would be equally unrealistic and would additionally introduce an unwarranted degree of complexity.

Under the above assumption, one can define two independent Damkohler numbers, one for the solute and one for the volatile as follows:

$$Da_s = \frac{\theta_{ga}^2}{\theta_r^2} \frac{C_{pg}}{\lambda_{gW_o}} \left(\frac{R^o C_{pg} \bar{W}}{R_u C_s} \right)^2 R_2^4 C^{-2} \exp \left(\frac{-\theta_r}{\theta_{ga}} \right) \times \begin{cases} Y_{oa} \text{ if oxygen-rich} \\ Y_{Fva,s} \phi'_{s,s} \text{ if oxygen-lean} \end{cases} \quad (1)$$

$$Da_v = \frac{\theta_{ga}^2}{\theta_r^2} \frac{C_{pg}}{\lambda_{gW_o}} \left(\frac{R^o C_{pg} \bar{W}}{R_u C_v} \right)^2 R_2^4 C^{-2} \exp \left(\frac{-\theta_r}{\theta_{ga}} \right) \times \begin{cases} Y_{oa} \text{ if oxygen-rich} \\ Y_{Fva,v} \phi'_{s,v} \text{ if oxygen-lean} \end{cases} \quad (2)$$

The symbols are all explained in the Nomenclature.

These Damkohler numbers are a measure of the importance of chemical rates with respect

to the diffusion rates, and have been used in the past to determine ignition of clusters of drops [17,20]. According to Law and Chung [20], ignition of a drop in a reactive environment will occur if the Damkohler number is larger than the ignition Damkohler number. The ignition Damkohler number is found [20] using large activation energy asymptotics in the quasi - steady gas phase equations for a single drop in surroundings where fuel vapor may be present. For a binary fuel, the ignition Damkohler numbers for solute and volatile are respectively obtained as explained in Bellan and Harstad [19]. For an oxygen-lean mixture, the correlation of Law and Chung [19] is used for Da_{ign} versus κ for $1 < \kappa < 10^3$. For oxygen-rich mixtures, the results presented in curve form by Law and Chung [19] for $0 < \gamma < 10^3$ and $0.005 < \beta < 1$ are recorelated by including values of Da_{ign} for $\gamma > 10^3$ and values of $\beta < 0.005$; these values are obtained by using asymptotic limits and matching by continuity.

Thus, ignition will be initiated by the solute if

$$Da_s \geq Da_{s,ign} \quad (3)$$

and ignition will be initiated by the volatile if

$$Da_v \geq Da_{v,ign} \quad (4)$$

When neither of these inequalities is satisfied during evaporation, the cluster of drops does not ignite. Limiting cases are ignition of drops just after they have completely evaporated, in which case $R_1 = 0.05$, or ignition of drops just as they have been introduced into the hot environment, in which case R_1 is very close to unity.

The model developed by Bellan and Harstad [19] not only predicted the timing of ignition, but also whether ignition occurs around isolated drops in a spray, around clusters of drops, or

inside clusters of drops but around a few of the drops rather than around the entire cluster. The criteria previously developed will be used here as well in order to determine the location of ignition.

RESULTS AND DISCUSSION

As mentioned above, the consensus from isolated drop studies is that ignition of binary - fuel drops is dominated by ignition of the volatile, and thus that by adding a small amount of volatile, ignition can be considerably enhanced. One of the goals of this study is to ascertain if this conclusion can be extended to a real spray which contains clusters of drops of various equivalence ratios, and thus of various number densities.

1. Typical behavior.

For the baseline set of solutions the solute was chosen to be No. 2 GT fuel oil (light Diesel oil) and the volatile was chosen to be n-decane. The initial values of the dependent variables are $u_d^0 = 200$ cm/s, $R_{cl}^0 = 3$ cm, $R_d^0 = 2 \times 10^{-3}$ cm, $T_{ga}^0 = 1000$ K, $T_{gs}^0 = 350$ K, $p = 1$ atm, $Y_{Fva}^0 = 0$, $Y_{HV,c}^0 = 2 \times 10^{-2}$. The properties of the two compounds are listed in [15], and the vapor pressure curves are presented there as well. The ignition chemical kinetic parameters where here chosen to be identical for the two compounds: $A_{ign} = 3.8 \times 10^{11}$ cm³/(mole-s), and $E_{ign} = 30$ kcal/mole [15,19]. Calculations were performed for an extended range of air/fuel mass ratios from 0.314 to over 25 (the stoichiometric value is 15) corresponding to a nondimensional radius of the sphere of influence of 8.4 to 37, thus encompassing the very dense, dense, dilute and very dilute regimes according to the definition given in Ref. 15.

The results show that for these initial conditions clusters having $\Phi^0 = 0.314$ do not ignite,

whereas clusters having $\Phi^0 = 0.785$ ignite at the time when the drops have completely evaporated (chosen to correspond to $R_1 = 0.05$). At the other extreme, for $\Phi^0 = 25$ the drops ignite at $R_1 = 0.98$, which is practically when they are introduced into the hot ambient. The ignition location is always outside the cluster, except for values of Φ^0 larger than 34, where ignition is found to occur inside the cluster at locations such that the resulting flame surrounds groups of drops. In all following calculations discussed in this paper, ignition occurred around clusters of drops. Figure 1 shows the ignition time, t_{ign} , versus R_2^0 , whereas Fig. 2 shows both t_{ign} and $R_{1,\text{ign}}$ versus Φ^0 . On the plots depicted in these figures, a distinction is made between situations when ignition occurs due to the solvent, and when ignition is due to the volatile.

The very dense regime is dominated by solvent ignition, whereas the very dilute regime is dominated by volatile ignition in agreement with the results of previous studies of isolated drops. These results are not surprising since it has been previously found [15] that evaporation is dominated by the solvent in the very dense regime whereas it is dominated by the volatile in the very dilute regime. Surprisingly, between these two extreme regimes ignition seems to be sometimes initiated by the volatile and sometimes by the solvent. The previous study of binary-fuel drop evaporation in clusters [15] does not provide an explanation to this surprising behavior.

In order to explain the behavior in the dense and dilute regimes, another set of calculations were performed where $A_{\text{ign},s}$ was taken negative if in the initial calculations it was found that the solvent was initiating ignition, and $A_{\text{ign},v}$ was taken negative if in the initial calculations it was found that the volatile was initiating ignition. Thus, ignition of the initiating compound was "turned off" in order to see if the other compound would ignite the cluster; and if ignition

occurred, the important consideration was to quantify the change in t_{ign} and $A_{1,\text{ign}}$. Results from these calculations show that in the very dense regime ignition does not occur at all, thus confirming the conclusion that ignition is strongly controlled by the solvent. In the very dilute regime, physically incorrect results are obtained where γ becomes negative; this confirms the conclusion that it is the volatile which strongly controls ignition in the very dilute regime. Between these two regimes, ignition is obtained at about the same t_{ign} and $R_{1,\text{ign}}$ as with the original calculations; the ratio of t_{ign} and $R_{1,\text{ign}}$ with the values obtained in the original calculations is at most 1%. This proves that it is neither the solvent nor the volatile which controls ignition in these intermediary regimes, and that it is only the details of the calculation which decide upon the compound initiating ignition. Since these details can never be modeled in a totally accurate manner, it is impossible to identify the compound initiating ignition in these intermediary regimes.

Since solvent-controlled or volatile-controlled evaporation has been associated with the value of Be , it is instructive to think again in terms of processes occurring at different characteristic times. Here there are two competing processes determining the compound initiating ignition: the relaxation rate of the slip velocity and the rate at which the Damkohler number approaches the ignition Damkohler number for either one of the compounds. If the rate at which the Damkohler number approaches the ignition Damkohler number is much slower than the relaxation rate of the slip velocity, then Be becomes very large by the time ignition occurs and thus the solvent controls ignition. In contrast, if ignition occurs very fast with respect to the relaxation rate of the slip velocity, then Be remains relatively small and the volatile controls ignition. Plots of the slip velocity versus t and of Be versus t can be found respectively in Figs. 3 and 4

corroborating the above interpretation.

The practical conclusion from these results is that in realistic spray situations where the system has been optimized for maximum efficiency one should never plan to control ignition by increasing the amount of volatile in the fuel. This is because the volatile strongly controls ignition only in the very dilute part of a spray, well past the point of efficient operation. Since usually it is the volatile which is the most expensive part of the fuel, this conclusion is important in reducing energy costs associated with using additional amounts of volatile in binary fuels. Rather than changing the composition of the fuel, it will be shown below that by changing the regime of operation, ignition can be made to be strongly controlled by the solvent in part of the dense regime adjacent to the very dense regime, and that the volatile can be made to strongly control ignition in parts of the dilute regime. Thus, at fixed composition one may control ignition using the solvent or the volatile by changing the surroundings of the spray.

2. Effect of the initial amount of volatile in the liquid.

Calculations were performed by increasing $Y_{HV,c}^0$ from 0.02 to 0.05 and 0.2. The results show identical values for t_{ign} and $R_{1,ign}$ in the very dense regime, in agreement with the conclusion that the volatile does not control ignition in this regime. In the very dilute regime, small changes were observed for both t_{ign} and $R_{1,ign}$, however, for these larger values of $Y_{HV,c}^0$ ignition occurs around the entire cluster rather than around groups of drops inside the cluster. For these larger amounts of volatile the amount of volatile vapor is correspondingly larger, and this stronger flux pushes the ignition location outside the cluster. The situation is similar to that of the flame motion for isolated drop evaporation: a stronger mass flux pushes the flame further away from the drop.

3. Effect of the volatile.

Calculations were performed by replacing n-decane by n-hexane and also by n-heptane. Both compounds have a larger latent heat than n-decane (more difficult to evaporate) and a higher saturation pressure (easier to evaporate). N-hexane has a larger latent heat than n-heptane and a higher saturation pressure than n-heptane. In all calculations $Y_{HV,c}^0 = 0.2$.

Results show negligible differences among the values for t_{ign} and $R_{1,ign}$ for all these runs. Obviously, in the very dense regime no differences are expected since the volatile is not controlling. What the results indicate is that in the very dilute regime the effect of the latent heat balances that of the saturation pressure, yielding no noticeable effect of the volatile. Note that in the model it is assumed that the volatility of the volatile is infinite with respect to that of the solvent, however differences among volatile properties are taken into account, so that if there were any noticeable effect of the volatile it would have been indicated in the results.

4. Effect of the solvent.

In order to explore the influence of the solvent, No. 2GT was replaced by n-decane and the volatile was chosen to be n-hexane. N-decane has a larger latent heat than No. 2GT oil and a larger saturation pressure. The results show substantial differences between the behavior of the two binary fuels.

In the very dense regime, ignition no longer occurs for clusters having $\Phi^0 = 0.785$. This indicates that the effect of the latent heat dominates that of the saturation pressure when the drops are in very close proximity. This is easy to understand since for dense clusters evaporation is limited by the availability of heat to the drops. The larger latent heat exacerbates the situation further, and prevents ignition. In the dense regime, the larger latent heat delays ignition as

shown in Fig. 5 where t_{ign} and $R_{1,\text{ign}}$ are plotted versus Φ^0 . Our interpretation of the results is confirmed by the much larger value of Be at fixed Φ^0 in the dense regime. Accordingly, when the solvent is n-decane instead of No. 2 GT oil, the range of strongly solvent-controlled ignition extends to larger values of R_2^0 . This is because, as explained above, for a given Φ^0 in this range, a cluster has stronger dense characteristics since L is larger.

In the very dilute regime the behavior of the binary fuel is exactly the opposite to that in the very dense and dense regimes: here ignition occurs earlier in time because it is promoted by the larger saturation pressure. The evaporation of dilute clusters of drops is not limited by drop heating because the drops behave as if they were isolated, and instead it is the saturation pressure which determines volatility. Although ignition occurs earlier in time, it occurs later in the drop lifetime because evaporation is faster due to the higher saturation pressure.

The ignition behavior in the dilute regime combines both effects of the latent heat and the saturation pressure. As a result, ignition is sometimes promoted and sometimes hindered as clearly seen in Fig. 5 .

5. Effect of the ambient temperature.

Decreasing the ambient temperature to 800K suppresses ignition in the very dense regime and delays ignition in all other regimes as shown in the plots of Fig. 6. The results show not only a loss of the strongly solvent-controlled ignition regime, but also a much milder control of the volatile in the very dilute regime. This is the consequence of a much slower evaporation which allows enough time for the relaxation of the slip velocity, yielding values of Be larger by at least one order of magnitude in the very dilute regime. Since $Be = O(10)$ as illustrated in Fig. 7, both liquid mass diffusion and surface layer stripping are important. Thus, decreasing

the ambient temperature not only suppresses ignition in the very dense regime and delays ignition for all Φ^0 's, but also results in a loss of control over ignition by either one of the two components. Therefore, at low ambient temperatures, ignition cannot be controlled in any regime by using the chemical constituents of the fuel.

A larger ambient temperature (1200K compared with 1000K) considerably enhances ignition to the point that ignition now occurs for smaller values of Φ^0 (for example 0.314 for which $R_2^0 = 8.9$). The strongly solvent-controlled regime extends both toward smaller and larger values of Φ^0 than in the typical case. The larger ambient temperature affects ignition in the dilute regime as well, resulting in immediate ignition for clusters having $R_2^0 = 24.4$. The much shorter evaporation time does not allow relaxation of the slip velocity, and thus now the volatile strongly controls ignition in the dilute regime. Depicted in Fig. 6 are t_{ign} and $R_{1,\text{ign}}$ versus Φ^0 , and plots of Be for selected values for Φ^0 appear in Fig. 8. The very dilute regime is not considered here since calculations are stopped in the Φ^0 domain (Φ^0 is no longer increased) as soon as ignition occurs instantaneously after introduction of the cluster into the ambient. The conclusion is that at higher temperature it is easier to control ignition over a wider range of Φ^0 's, either using the solvent in the very dense and dense regimes, or using the volatile in the dilute regime. Since sprays contain clusters of drops having initially a range of air/fuel mass ratios, this means that ignition control of the *entire spray* can be achieved at high temperatures by using the chemical components of the fuel as drivers.

6. Effect of the chemical kinetics of ignition.

All the results obtained so far have contained the assumption that the ignition kinetic parameters of solvent and volatile are identical. Results from calculations where $E_{\text{ign},s}$ has been

increased by 3% show that ignition is suppressed in the very dense regime, whereas in all other regimes there are only minor changes with respect to the baseline results (now there is an apparent domination of the volatile for the entire range of Φ^0 's, except in a very narrow range in the dense regime). Thus, a small change in the activation energy of ignition results in loss of the strongly solvent-controlled ignition regime.

A 3% increase in $E_{\text{ign},v}$ does not produce any changes in the results obtained in the very dense regime, however ignition does no longer occur in the very dilute regime. This is an additional indication that the volatile strongly controls ignition in the very dilute regime. For all other values of Φ^0 , there is an apparent solvent dominated ignition. Similar to the conclusion when changing $E_{\text{ign},s}$, now it is the control of the volatile which is lost.

This study thus shows that the ignition kinetics has an overwhelming effect on ignition, overshadowing all other parameters. For this reason, it is recognized that no definitive results can be obtained in this investigation until the values of the chemical kinetic parameters are better known.

7. Effect of the initial slip velocity.

It is here recalled that since the gas in the cluster is initially at rest, the initial slip velocity is the initial velocity of the drops. Reducing the initial slip velocity to 100 cm/s from the typical value of 200 cm/s, results in a decrease in the evaporation rate, and this generally slightly increases the ignition time, without much change in the value of $R_{1,\text{ign}}$. In the very dense regime where evaporation is hindered by drop proximity, the lowering of u_d^0 suppresses ignition. The smaller evaporation rate in the dense regime allows relaxation of the slip velocity, resulting again in solvent strongly-controlled ignition. The change in the initial slip velocity

affects the dense regime much less than the very dilute regime since in the dense regime the relaxation of the slip velocity is faster. In the very dilute regime the evaporation rate and the slip velocity relaxation rate are still comparable, so that ignition is still strongly-controlled by the volatile.

Thus the solvent strongly-controlled ignition regime moves into the dense regime whereas volatile strongly-controlled ignition remains in the very dilute regime.

8. Effect of the cluster initial radius.

When the initial radius of the cluster is decreased to be 1 cm instead of the baseline value of 3 cm, the surface/volume ratio increases accordingly. This enhances heat transfer processes from the ambient to the cluster. Heat transfer processes are very important in the dense regime, but are unimportant in the dilute regime where evaporation is not limited by the amount of heat available for each drop. As a result, ignition is now obtained for smaller values of Φ^0 (denser clusters) than in the corresponding calculation with the larger initial cluster radius as can be seen in Fig. 9. The solvent strongly-controlled ignition regime moves accordingly toward the smaller Φ^0 's; this is also confirmed by plots of Be versus R_i shown in Fig. 10. The very dilute regime is unaffected by the change in the initial cluster radius, as discussed above.

SUMMARY AND CONCLUSIONS

A model of binary-fuel drop evaporation in clusters has been used in conjunction with an ignition model based upon the Damkohler number criterion for drops in sprays to study ignition of clusters of drops. The assumption is also made that the chemistry of the two compounds is independent from each other.

Calculations were performed by varying the air/fuel mass ratio for different values of the main parameters. Small values of the air/fuel mass ratio correspond to very dense clusters of drops, whereas large values of the air/fuel mass ratio correspond to very dilute clusters of drops in which there is no drop interaction. Baseline results show that in a typical ambient gas temperature, ignition is strongly controlled by the solvent in the very dense cluster regime, and that ignition is strongly controlled by the volatile in the very dilute regime. In the intermediary dense and dilute regimes, control by either one of the compounds is very weak and depends entirely upon the details of the calculation. Thus, the isolated drop results are recovered in the very dilute regime, however it is found that it is qualitatively a different process which controls ignition in a real spray. The practical conclusion is that at typical ambient gas temperatures it is useless to use more expensive fuels having larger amounts of volatile for the purpose of enhancing ignition because it is only the very dilute clusters of drops which will benefit from this treatment. These clusters of drops appear only at relatively large distances from the atomizer and thus do not participate in the ignition process.

As the ambient temperature of the gas is increased, it is easier to gain control of ignition over a larger range of air/fuel mass ratios. This is achieved by using the volatile in the very dilute and increasingly in the dilute regime, and by using the solvent in the very dense and increasingly in the dense regime. Eventually, a temperature is reached at which ignition control of the entire spray can be achieved through ignition control of clusters at all air/fuel mass ratios.

Studies of the effect of the compound used as solvent revealed that the concept of volatility is associated more with the latent heat of evaporation in the very dense cluster regime and is associated more with the saturation pressure in the very dilute cluster regime.

It is also shown that variations of the initial slip velocity between phases affect the results only in the very dilute regime because of the short relaxation time of the slip velocity in the very dense regime. In contrast, changes in the size of the cluster affect the ignition timing only in the very dense regime which is limited by the amount of heat available to each drop to increase its temperature and evaporate.

For very low values of the initial amount of volatile in the liquid, ignition in the very dilute regime is found to occur around groups of drops inside the cluster. For air/fuel mass ratios outside the very dilute regime ignition occurs around the entire cluster of drops. As the initial amount of volatile in the liquid is increased, it is found that ignition occurs exclusively around the entire cluster.

All of the above results were found under the assumption that the chemical kinetic parameters of the two compounds were identical. Studies of the effect of the chemical kinetic parameters show that these have an overwhelming effect on the ignition results. Solvent-controlled ignition can be lost when the solvent activation energy is slightly increased. Equivalently, volatile-controlled ignition can be lost when the volatile activation energy is slightly increased. Since chemical kinetic parameters of hydrocarbons are not known for hydrocarbons more complicated than propane, definitive conclusions must await precise chemical kinetic information.

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NOMENCLATURE

A	preexponential factor in the Arrhenius rate
Be	$-(R/D_m u_l)^{0.5} dR/dt$
C	nondimensional evaporation rate, $-m/[4 \pi (\rho_g D)^\infty R^0]$
C	heat of combustion
C_p	heat capacity at constant pressure
D	gas phase diffusivity
Da	Damkohler number
D_m	coefficient of mass diffusion in the liquid
E	activation energy in the Arrhenius rate
L	latent heat of evaporation
\mathcal{L}	effective heat of evaporation, $L + 4\pi R_2 \lambda_l (\partial T_l / \partial r)_{r=R_2} / m$
m	evaporation rate
p	pressure
r	radial coordinate
R	drop radius
R_1	R/R^0
R_2	nondimensional radius of the sphere of influence
R_u	universal gas constant
t	time
T	temperature
u	velocity

u_i	velocity of the vortex motion inside the drop
w	molecular weight
Y	mass fraction

Greek Symbols

β	$[(\theta_{ga} - \theta_{gs})L_{bn}/C + 1] \mathcal{L}/C$
γ	$Y_{Fva}\theta_r C/(\theta_{ga}L_{bn})$
θ	$C_p T/L_{bn}$
θ_r	$C_{pg}E/R_u L_{bn}$
κ	$Y_{Fva}\theta_r C/(\theta_{ga}^2 L_{bn} \Phi_s)$
λ	conductivity
Φ (Φ_s)	air/fuel mass ratio (stoichiometric)
Φ' (Φ'_s)	oxygen/fuel mass ratio (stoichiometric)
ξ	self-similarity variable

Subscripts

a	ambient; edge of the sphere of influence
bn	normal boiling point
c	core of the drop
cl	cluster
d	drop
e	cluster edge

Fv fuel vapor
g gas
HV high volatility
ign ignition
s solvent
sl slip
sf drop surface
v volatile

Superscripts

0 initial
— average

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FIGURE CAPTIONS

- Figure 1. Ignition time versus the initial nondimensional radius of the sphere of influence. Initial conditions are: $u_d^0 = 200$ cm/s, $R_{cl}^0 = 3$ cm, $R_d^0 = 2 \times 10^{-3}$ cm. $T_{gs}^0 = 1000$ K, $T_{ga}^0 = 350$ K, $p = 1$ atm, $Y_{Fva,C}^0 = 0$, $Y_{HV,C}^0 = 2 \times 10^{-2}$. The solvent is No. 2 GT fuel oil and the volatile is n-decane. S designates solvent ignition and V designates volatile ignition.
- Figure 2. Ignition time and residual drop radius versus the initial air/fuel mass ratio for the same conditions as those of Fig. 1. Filled triangles designate solvent ignition and white triangles designate volatile ignition.
- Figure 3. Slip velocity versus time for selected values of the initial air/fuel mass ratio for the same conditions as those of Fig. 1. $\Phi^0 = 0.785$ (o), $\Phi^0 = 1.57$ (Δ), $\Phi^0 = 3.14$ (\square), $\Phi^0 = 7.85$ (\diamond), $\Phi^0 = 25.12$ (∇)
- Figure 4. Ratio of the drop regression rate to a characteristic volatile diffusion rate versus time for selected values of the initial air/fuel mass ratio for the same conditions as those of Fig. 1. The symbols are the same as in Fig. 3.
- Figure 5. Ignition time and residual drop radius versus the initial air/fuel mass ratio for two binary fuels having hexane as the volatile with $Y_{HV,C}^0 = 2 \times 10^{-1}$. In one case the solvent is No.2 GT fuel oil (\bullet and \square), in the other case the solvent is n-decane (Δ and \diamond). The other initial conditions are those of Fig.1.
- Figure 6. Ignition time and residual drop radius versus the initial air/fuel mass ratio for a binary fuel composed of No. 2 GT oil and n- decane. The solvent is No. 2 GT oil and the volatile is n-decane with $Y_{HV,C}^0 = 2 \times 10^{-1}$. The values of T_{gs}^0 are 800K (Δ , ∇), 1000K (\bullet , \diamond), and 1200K (\blacksquare , O). The other initial conditions are those of Fig. 1.
- Figure 7. Variation of Be versus the residual drop radius for selected values of Φ^0 and for the same initial conditions as in Fig. 6 with $T_{gs}^0 = 800$ K. $\Phi^0 = 3.14$ (∇), $\Phi^0 = 4.7$ (\diamond), $\Phi^0 = 5.4$ (\blacksquare), $\Phi^0 = 7.85$ (Δ), $\Phi^0 = 25.12$ (\bullet).
- Figure 8. Variation of Be versus the residual drop radius for selected values of Φ^0 and for the same initial conditions as in Fig. 6 with $T_{gs}^0 = 1200$ K. $\Phi^0 = 0.314$ (∇), $\Phi^0 = 0.785$ (\diamond), $\Phi^0 = 1.57$ (\square), $\Phi^0 = 3.14$ (Δ), $\Phi^0 = 6.3$ (\bullet).
- Figure 9. Ignition time and residual drop radius versus the initial air/fuel mass ratio for a cluster with $R_{cl}^0 = 3$ cm (\bullet , \blacksquare) and a cluster with $R_{cl}^0 = 1$ cm (Δ , \diamond). The initial conditions are those of Fig. 1 except that $Y_{HV,C}^0 = 2 \times 10^{-1}$.
- Figure 10. Variation of Be versus R_1 for selected values of Φ^0 when $R_{cl}^0 = 1$ cm. The other initial conditions are those of Fig. 9. $\Phi^0 = 0.314$ (o), $\Phi^0 = 0.785$ (∇), $\Phi^0 = 1.57$ (\diamond), $\Phi^0 = 3.14$ (\blacksquare), $\Phi^0 = 7.85$ (Δ), $\Phi^0 = 25.12$ (\bullet).











